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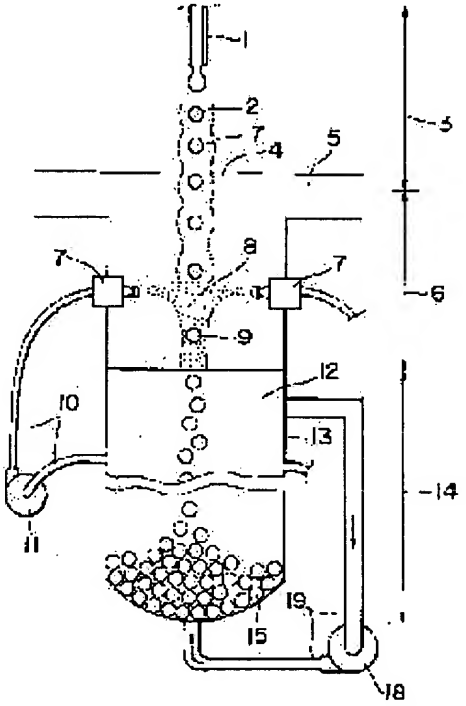
(54) PROCESS AND APPARATUS FOR PRODUCTION OF AMMONIUM DIURANATE PARTICLE

(57)Abstract:
PURPOSE: To easily produce ammonium diuranate having high sphericity with a small-sized device by spraying the mist of an aq. ammoniacal soln. to the liquid drops of an uranyl nitratesol.
CONSTITUTION: The liquid drops 2 of the ammonium diuranate soln. are dropped by vibration from a dropping nozzle 1. A liquid drop forming section 3 has the distance sufficient for the liquid drops 2 to be made spherical. The gaseous ammonia is sufficiently discharged from a discharge port 5 so as to prevent the leakage of the gas to the liquid drop forming section 3. The mists (≤30μm average diameter) 8 of the aq. ammoniacal soln. (e.g.: satd. ammonia water) are sprayed to the liquid drops 2 entering a surface gelatinizing section 6. The liquid drops 9 having the gelatinized surfaces arrive at the liquid surface of the aq. ammoniacal soln. 12 which is a complete gelatinizing section 14. While the liquid drops fall in the aq. ammoniacal soln. 12 in a setting chamber 13, the surfaces of the liquid drops 9 are completely gelatinized to form the ammonium diuranate particles 15. A pump 11 feeds the liquid and a circulating pump 18 circulates the settling chamber 13.



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* NOTICES *

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CLAIMS

[Claim(s)]
[Claim 1] The manufacture approach of the heavy uranic acid ammonium particle characterized by spraying Myst of an ammonia nature water solution on the drop of an uranyl-nitrate solution in the approach of manufacturing a heavy uranic acid ammonium particle, by the external gelling method from an uranyl-nitrate solution.
[Claim 2] The manufacture approach of said heavy uranic acid ammonium particle according to claim 1 that said ammonia nature water solution is saturated ammonia water.
[Claim 3] The manufacture approach of said heavy uranic acid ammonium particle according to claim 1 that said Myst is formed with the sprayer of an ultrasonic sensing method.
[Claim 4] Said Myst is the manufacture approach of said heavy uranic acid ammonium particle according to claim 3 that the average diameter is 30 micrometers or less.
[Claim 5] In the heavy uranic acid ammonium particle manufacturing installation which manufactures a heavy uranic acid ammonium particle by the external gelling method from an uranyl-nitrate solution It has a drop dropping means by which an uranyl-nitrate solution is dropped, and the setting tank which has the ammonia nature water solution which receives the drop of the uranyl-nitrate solution dropped from said drop dropping means. Between said drop dropping means and said setting tanks The heavy uranic acid ammonium particle manufacturing installation characterized by coming to prepare the atomizer which sprays said Myst of an ammonia nature water solution.
[Claim 6] Said heavy uranic acid ammonium particle manufacturing installation according to claim 5 which comes to have a means to send an ammonia nature water solution from said setting tank to said atomizer.

[Translation done.]

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Industrial Application] This invention relates to the heavy uranic acid ammonium particle manufacturing installation of the simple structure used for the manufacture approach of a heavy uranic acid ammonium particle that heavy uranic acid ammonium with high sphericity can be simply manufactured with small equipment in more detail, and its manufacture approach about the heavy uranic acid ammonium particle manufacturing installation used for the manufacture approach of a heavy uranic acid ammonium particle, and its manufacture approach.
[0002]
[Description of the Prior Art] Generally, a heavy uranic acid ammonium particle is manufactured by the external gelling method, as shown in drawing 2 . The drop 2 of the uranyl-nitrate solution formed by the drop dropping means 1 becomes a real ball with the viscosity of itself, and surface tension, while falling the drop formation section 3. Said drop used as a real ball falls from the drop fall opening 4 at the surface gelation section 6. Subsequently, said drop is completely gelled in the ammonia nature water solution which is the full gelation section, and it becomes a heavy uranic acid ammonium particle.
[0003] The sphericalness of a heavy uranic acid ammonium particle is governed by the sphericalness of the drop of the uranyl-nitrate solution which landed on the water into the ammonia nature water solution, and the sphericalness of the drop which deformed does not revert in the full gelation section. Therefore, the purpose of the surface gelation section is fully gelling the front face of said drop by ammonia gas so that an impact when the drop of an uranyl-nitrate solution lands on the water into an ammonia nature water solution may be borne and the sphericalness can be held.
[0004] As a general method of making a front face gel, the method of passing said drop is used into gelling agent ambient atmospheres, such as ammonia. As the concrete approach, conventionally, ammonia gas was blown as a jet into the layer of air from the ammonia gas supply opening 16, and the approach of promoting surface gelation of said drop was taken. On the usual drop formation conditions, the termination fall rate of a drop is about 0.5m/second or more. At this rate, since the falling drop incorporates air (or inert gas) to that perimeter in the drop formation section, as shown in drawing 2 , the layer 17 of air is formed in the perimeter of a drop 2 also in the surface gelation section. Since the layer of this air checks the reaction of a drop and ammonia gas, it needs to blow ammonia gas as a jet in the layer of air.
[0005] Since ammonia gas is supplied in the state of a jet, the first fault by the above-mentioned approach is that a drop deforms by this. Furthermore, since the amount of the ammonia gas used increases, it is that ammonia gas recovery equipment to prevent environmental pollution is enlarged, therefore the whole heavy uranic acid ammonium particle manufacturing installation is enlarged.
[0006] If the second fault lengthens distance of the surface gelation section in order to fully perform surface gelation of a drop, a termination fall rate in case a drop lands at an ammonia nature water solution will also increase, and the impact at the time of splashdown will become large. Therefore, even if it is gelling the front face of a drop enough by ammonia gas, it is that sphericalness will be destroyed with the impact at the time of splashdown.
[0007] This invention is aimed at solving said technical problem. That is, the purpose of this invention is offering the heavy uranic acid ammonium particle manufacturing installation used for the manufacture approach of a heavy uranic acid ammonium particle a heavy uranic acid ammonium particle with high sphericity being manufactured with small equipment, and its manufacture approach.
[0008]
[Means for Solving the Problem] In order to solve said technical problem, as a result of this invention persons' inquiring wholeheartedly, invention according to claim 1 In the approach of manufacturing a heavy uranic acid ammonium particle by the external gelling method from an uranyl-nitrate solution It is the manufacture approach of the heavy uranic acid ammonium particle characterized by spraying Myst of an ammonia nature water solution on the drop of an uranyl-nitrate solution. Invention according to claim 2 Said ammonia nature water solution is the manufacture approach of said heavy uranic acid ammonium particle according to claim 1 which is saturated ammonia water. Invention according to claim 3 Said Myst is the manufacture approach of said heavy uranic acid ammonium particle according to claim 1 formed with the sprayer of an ultrasonic sensing method. Invention according to claim 4 Said Myst is the manufacture approach of said heavy uranic acid ammonium particle according to claim 3 that the average diameter is 30 micrometers or less. Invention according to claim 5 In the heavy uranic acid ammonium particle manufacturing installation which manufactures a heavy uranic acid ammonium particle by the external gelling

method from an uranyl-nitrate solution It has a drop dropping means by which an uranyl-nitrate solution is dropped, and the setting tank which has the ammonia nature water solution which receives the drop of the uranyl-nitrate solution dropped from said drop dropping means. Between said drop dropping means and said setting tanks It is the heavy uranic acid ammonium particle manufacturing installation characterized by coming to prepare the atomizer which sprays said Myst of an ammonia nature water solution. Invention according to claim 6 It is said heavy uranic acid ammonium particle manufacturing installation according to claim 5 which comes to have a means to send an ammonia nature water solution from said setting tank to said atomizer.

[0009] Hereafter, it explains in more detail about this invention.

(1) An uranyl-nitrate solution uranyl-nitrate solution is prepared from an uranyl-nitrate undiluted solution, pure water, a thickener, etc. An uranyl-nitrate undiluted solution is U3 O8. It can obtain by dissolving powder in a nitric acid. The uranium concentration in an uranyl-nitrate undiluted solution is usually 465 to 475 gU/a liter.

[0010] Since the drop of the uranyl-nitrate solution with which the thickener was dropped comes real ball-like with the surface tension and viscosity of itself during fall, it is added. The resin which has as a thickener the property solidified under polyvinyl alcohol resin or alkali conditions, for example, a polyethylene glycol, METOROZU, etc. can be mentioned. as the resin which has the property solidified under alkali conditions -- resin -- even if independent, what has the coagulation engine performance in an alkali ambient atmosphere, for example, polyvinyl-acetal / N;N-dimethylamino acetate acid neutralization object etc., (trade name AEA, product made from Sankyo Pharmaceuticals) can be mentioned. The kind can also be independently used for a thickener, and it can also use together the two or more sorts.

[0011] The uranium concentration in an uranyl-nitrate solution is usually 120 to 250 gU/a liter preferably gU(s)/100 liters - 400 gU/a liter. Although it is 2g/l. - 50g/l. as a general content of a thickener, when using METOROZU as a thickener, the concentration is usually 4-10g/l. In addition, in this uranyl-nitrate solution, additives, such as a photolysis halt agent and a surfactant for preparing the surface tension of an undiluted solution, may be contained suitably. The matter which is effective in preventing the photolysis (uranium serving as a catalyst) of binder resin as a photolysis halt agent, for example, tetrahydro furil alcohol etc., (4HF) can be mentioned.

[0012] Although there is no limit especially in the preparation approach of an uranyl-nitrate solution, in the usual case, a thickener and pure water are mixed, a thickener water solution is prepared beforehand, this thickener water solution and uranyl nitrate are mixed, and, subsequently it is prepared by adding pure water as preparation of concentration or viscosity.

[0013] (2) By cooling to predetermined temperature, the viscosity is prepared and the uranyl-nitrate solution which is the formation approach above of an uranyl-nitrate solution drop, and was made and prepared is dropped at an aqueous ammonia solution from a dropping nozzle. At this time, the cooling temperature of the uranyl-nitrate solution dropped from a dropping nozzle is determined by how that viscosity is prepared. For example, if you are going to make it hold the viscosity of an uranyl-nitrate solution to 70 - 100cp (centipoise), it is good to make cooling temperature into 18-20 degrees C.

[0014] Especially the drop-ized approach of an uranyl-nitrate solution is realizable by vibrating a narrow diameter dropping nozzle with a proper means, for example, although there is no limit. In case a dropping nozzle is vibrated, even if it vibrates the shaft orientations to a dropping nozzle, you may make it vibrate in the diameter direction of a dropping nozzle. Although vibration frequency is 40-200Hz, its about 150Hz is usually the most common. As a path of a dropping nozzle, 0.4-1.5mm can usually be mentioned. The amount of sending out of the uranyl-nitrate solution sent out from a dropping nozzle is usually a part for 15-30 cc/. The drop of an uranyl-nitrate solution which has abbreviation and a 1.2-2.8mm diameter on such dropping conditions of an uranyl nitrate is formed of a dropping nozzle. A dropping nozzle is arranged so that the aqueous ammonia solution later mentioned while turning the opening caudad may be attended.

[0015] (3) In space until it lands at the aqueous ammonia solution front face of the setting tank which has the aqueous ammonia solution mentioned later, the drop of the uranyl-nitrate solution formed by vibration of the surface gelation approach dropping nozzle of an uranyl-nitrate solution drop needs to fully gel the front face of a drop in order to prevent the deformation at the time of splashdown. However, since it is not yet a real ball and an oscillating nozzle tip blockades in an instant if an oscillating nozzle tip contacts ammonia gas, as for the drop immediately after [an oscillating nozzle to] fall, it is desirable to make it the ambient atmosphere of inert gas, such as air or nitrogen, from a dropping nozzle in the fixed space turned caudad.

[0016] This invention makes Myst of an ammonia nature water solution spray on a drop as a means to make the front face of a drop gel. Although there will be no limit as an ammonia nature water solution which is a gelling agent especially if ammonia gas is generated, aqueous ammonia or the heated hydrazine water solution is desirable, and especially saturated ammonia water is suitable. The concentration of an aqueous ammonia solution is usually 20 % of the weight - saturated concentration.

[0017] Being incorporated in the layer of air or inert gas, and generating ammonia gas, Myst of an ammonia nature water solution adheres to a drop, and gels the front face of a drop. A gel front face is formed when a heavy uranic acid solution converts into an ammonium compound [like heavy uranic acid Amon (ADU)], whose metal salt, for example, uranyl nitrate, are, by the reaction with ammonia. Myst is far more efficient rather than a surface gelation operation of this drop is based only on ammonia gas.

[0018] The diameter of Myst is so good that it is small. This is for the ammonia gas which the direction of small Myst adhering to a drop front face at homogeneity and the specific surface area of Myst become large, and evaporates from Myst to increase. If the diameter of Myst is about 30 micrometers or less, it is fully effective.

[0019] As a sprayer made to generate Myst, all of the sprayer of a compressed-gas method and the sprayer of an ultrasonic sensing method can be used. By the compressed-gas method, a drop may deform by the jet of compressed gas. On the other hand, when the sprayer of an ultrasonic sensing method is used, the nozzle point which oscillates a supersonic wave has some generation of heat. For this reason, since much ammonia gas will evaporate out of Myst if an ammonia nature water solution is supplied to the atomizer of an ultrasonic sensing method, the effectiveness of a surface gelation operation becomes still better. Therefore, in this invention, an ultrasonic-type sprayer is used more suitably.
[0020] It is usually 3 times the flow rate of the ammonia nature water solution sprayed as Myst of this from 1 time to the amount of drops per oscillating nozzle, and is it from twice preferably. [of this] [2.5 times] When [than 3 times] more [the sphericalness of a drop cannot fully secure if there are few amounts of Myst than 1 time, and], there are many losses of Myst.
[0021] By the approach of this invention, although there is no deformation of the drop by the surface gelation means, the reason is as follows. The layer of air or inert gas is flowing caudad at the almost same rate as a drop. For this reason, Myst incorporated by the layer of air or inert gas also flows below with the flow of air or inert gas. That is, since it passes through the location of an atomizer and a drop falls with the vapor-liquid two-phases flow of ammonia gas and Myst of an ammonia nature water solution, there is no deformation by surface gelation.
[0022] (4) **** by which the setting tank front face which has an ammonia nature water solution was gelled is gelled completely, landing at the ammonia nature water solution stored in the setting tank, and sedimenting toward the bottom of a setting tank. The same thing as what was used when generating Myst of the aqueous ammonia solution in a setting tank is desirable. It is because the cyclic use of waste water that pump out of a setting tank using a duct and a pump, spray as Myst, and Myst is collected in a setting tank becomes possible.
[0023] thus, the heavy uranic acid ammonium particle completely gelled in the setting tank should be washed with pure water, ethanol, or a methanol, and pass desiccation, roast, and a sintering stroke further -- spherical -- UO2 It becomes a nucleus fuel particle. These processings of each can be performed by the well-known approach.
[0024] (5) The heavy uranic acid ammonium particle manufacturing installation of heavy uranic acid ammonium particle manufacturing installation this invention is for enforcing the manufacture approach explained above. The outline of equipment is shown in drawing 1 . The following is explained referring to drawing 1 .
[0025] As shown in drawing 1 , a heavy uranic acid ammonium particle manufacturing installation is equipped with a drop dropping means and a setting tank 13. Said drop dropping means is equipped with the dropping nozzle 1 which trickles the uranyl-nitrate solution supplied from the uranyl-nitrate solution feed zone which the place which stores an uranyl-nitrate solution does not illustrate. The dropping nozzle 1 is perpendicularly arranged [above said setting tank 13] while having opening which carries out opening caudad. Vibration can be horizontally given by the proper means and this dropping nozzle 1 can drop now the drop adhering to that opening.
[0026] A setting tank 13 makes the tube-like object of the owner bottom which has the drop fall opening 4 in the upper part. In this setting tank 13, only the amount predetermined in the ammonia nature water solution 12 is stored. It is a circumferential side face in a setting tank 13, and is the drop fall opening 4 bottom, and rather than the oil level in a setting tank 13, the exhaust port 5 of a pair carries out phase opposite, and is established in the upper part. The pipe which collects ammonia gas with the displacement of extent which does not disturb an ambient atmosphere in case a drop 2 falls is connected to this exhaust port 5. In addition, said pipe is equipped with the exhaust air pump which is not illustrated in order to exhaust ammonia gas through a pipe.
[0027] The atomizer 7 which sprays Myst of an ammonia nature water solution on a drop between said exhaust ports 5 and oil levels of the ammonia nature water solution 12 in a setting tank 13 is installed in the circumferential side face of a setting tank 13. In addition, if this atomizer 7 is in the equipment shown in this drawing 1 , it makes Myst generate using a supersonic wave, although what was explained by the term of "the formation approach of (2) uranyl-nitrate solution drop" can be suitably used for it. The duct 10 is attached in the wall surface of the part which is in contact with the ammonia nature water solution of a setting tank 13, and the ammonia nature water solution 12 can be supplied now to an atomizer 7 with a pump 11. Moreover, the duct 19 is attached so that the wall surface of the upper part of the part which is in contact with the pars basilaris ossis occipitalis and ammonia nature water solution of a setting tank 13 may be connected, and it circulates through an ammonia nature water solution upwards from a pars basilaris ossis occipitalis with the ammonia nature water-solution circulating pump 18.
[0028] If it is in the equipment shown in this drawing 1 , from the dropping nozzle 1 to about four drop fall opening is the drop formation section 3, from about four drop fall opening to the ammonia nature water-solution side 12 is the surface gelation section 6, and the inside of the ammonia nature water solution 12 serves as the full gelation section 14.
[0029] The operation of a heavy uranic acid ammonium particle manufacturing installation shown in drawing 1 is explained below. First, the drop 2 of a heavy uranic acid ammonium solution is trickled by vibration from the dropping nozzle 1. Since the drop formation section 3 has sufficient distance for a drop 2 to become a real ball and is arranged in air or an inert gas ambient atmosphere, even if the drop 2 immediately after fall does not serve as a real ball, a drop 2 will become a real ball, falling the drop formation section 3. Sufficient exhaust air is performed from the exhaust port 5 so that ammonia gas may not be revealed from the drop fall opening 4 at the drop formation section 3. Therefore, there is no deformation of the drop configuration by ammonia gas acting on the drop which is not a real ball, and making a front face gel in the drop formation section 3 also by this. Moreover, ammonia gas acts on opening of a dropping nozzle, and the opening does not blockade.
[0030] Myst 8 of an ammonia nature water solution is sprayed on the drop 2 which went into the surface gelation

section 6 from the drop fall opening 4 from an atomizer 7. Since 2 machine wearing is carried out, an atomizer 7 can spray Myst 8 on homogeneity over the whole drop 2. Subsequently, the drop 9 by which the front face was gelled lands at the oil level of the ammonia nature water solution 12 which is the full gelation section 14. Since the front face of a drop 2 is gelled by Myst 8 of said ammonia nature water solution at this time, a drop is not damaged at the time of splashdown. Falling the inside of the ammonia nature water solution 12 in a setting tank 13, the drop 9 by which the front face was gelled is gelled completely, and serves as the heavy uranic acid ammonium particle 15.

[0031] In this invention, the ammonia nature water solution 12 in a setting tank 13 is extracted, and the liquid is sent to an atomizer 7 with a duct 10 and a pump 11. Myst 8 of the sprayed ammonia nature water solution is collected by the setting tank 13. Therefore, since this invention does not newly need to supply ammonia gas like the conventional approach, it can miniaturize the ammonia gas recovery equipment connected to the point of an exhaust port 5. Moreover, since the ammonia nature water solution in a setting tank 13 is circulated upwards from the pars basilaris ossis occipitalis with the ammonia nature water-solution circulating pump 18, it can prevent the heavy uranic acid ammonium particle's 15 piling up at the pars basilaris ossis occipitalis of a setting tank 13, and deforming by the self-weight.

[0032] In addition, the die length of the drop formation section 3, the surface gelation section 6, and the full gelation section 14, the location of an atomizer 7, etc. can be suitably defined by experiment in consideration of the magnitude of a drop, a diameter, an amount of Myst, etc.

[0033] The heavy uranic acid ammonium particle manufacturing installation of this invention is not limited to what was shown in drawing 1 , and can carry out a design change variously within the limits of the summary of this invention. For example, two or more drop dropping nozzles may be prepared, and the number of atomizers may be increased in connection with it.

[0034]

[Example] Next, although the example of this invention is shown, this invention is not limited at all.

[0035] (Example 1) The uranyl-nitrate solution of viscosity 92cp (30 degrees C) was prepared with the 30g [/l.] polyvinyl alcohol resin as the uranium concentration of 250g/l., and a high molecular compound. The drop with a diameter of 2.1mm was made to form using this uranyl-nitrate solution on conditions with a vibration frequency [of an oscillating nozzle / of 100Hz], and an amplitude of 0.36mm.

[0036] Subsequently, this drop was dropped at the surface gel section which sprayed Myst on condition that the following. Myst was generated using the ultrasonic type atomizer (the product made from SONOTEKKU, nozzle bore:1.32mm, a frequency: 120kHz) in the service condition of amount of liquid sending 30 cc/min of an ammonia nature water solution. It was 18 micrometers in average diameter of Myst. Since two sets of ultrasonic atomizers were used, the total Myst yield was 60 cc/min.

[0037] The drop fell the distance of 35cm from an oscillating nozzle lower limit to the water surface of an ammonia nature water solution, and the termination fall rate at the time of splashdown was 3.5m/second. The drop of this example also bore the impact at the time of splashdown enough, and maintained the sphericalness. Furthermore, the sphericalness was able to be held, even when the formation conditions of a drop were changed and a termination fall rate was made high to 8.7m/second. The tolerance of the termination fall rate in the case of blowing ammonia gas as a jet and making the front face of a drop gel was usually about 4m/second. This result shows that the effectiveness which the way of the approach using Myst of an ammonia nature water solution makes gel the front face of a drop is good rather than the approach by conventional ammonia gas.

[0038] The sphericalness was evaluated after washing the heavy uranic acid ammonium particle manufactured using the heavy uranic acid ammonium particle generation equipment of this invention by pure water and ethanol. Sphericity was estimated by the following technique. That is, the diameter of one particle is measured 50 times at random using a particle size analyzer. The ratio of the max of this diameter measured value and min expresses sphericity (sphericity = a diameter at the maximum equator / the minimum diameter). That is, a geometric real ball is set to 1. The comparison of the sphericity of this invention and the particle of a conventional method is shown in Table 1.

[0039]

[Table 1]

平均直径（mm）	本発明の真球度	従来法の真球度
2.12	1.03	1.08
1.81	1.05	1.10
1.53	1.04	1.09

[0040] Furthermore, by the approach of this invention, since it is not necessary to newly supply ammonia gas in the surface gelation section, the throughput of ammonia gas recovery equipment to prevent environmental pollution can be reduced to about 40%.

[0041]

[Effect of the Invention] According to this invention, the heavy uranic acid ammonium particle manufacturing installation used for the manufacture approach of a heavy uranic acid ammonium particle that a heavy uranic acid ammonium particle with high sphericity can be manufactured with small equipment, and its manufacture approach can be offered.

[0042]

[Translation done.]

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]
[Drawing 1] Drawing 1 is the explanatory view of the manufacturing installation of the heavy uranic acid ammonium particle by this invention.
[Drawing 2] Drawing 2 is the explanatory view of the manufacturing installation of the heavy uranic acid ammonium particle by the conventional external gelling method.
[Explanation of agreement]
1 Dropping Nozzle
2 Drop
3 Drop Formation Section
4 Drop Fall Opening
5 Exhaust Port
6 Surface Gelation Section
7 Atomizer
8 Myst of Ammonia Nature Water Solution
9 Drop by Which Front Face was Gelled
10 Duct
11 Pump
12 Ammonia Nature Water Solution
13 Setting Tank
14 Full Gelation Section
15 Heavy Uranic Acid Ammonium Particle
16 Ammonia Gas Supply Opening
17 Layer of Air
18 Ammonia Nature Water-Solution Circulating Pump
19 Duct

[Translation done.]

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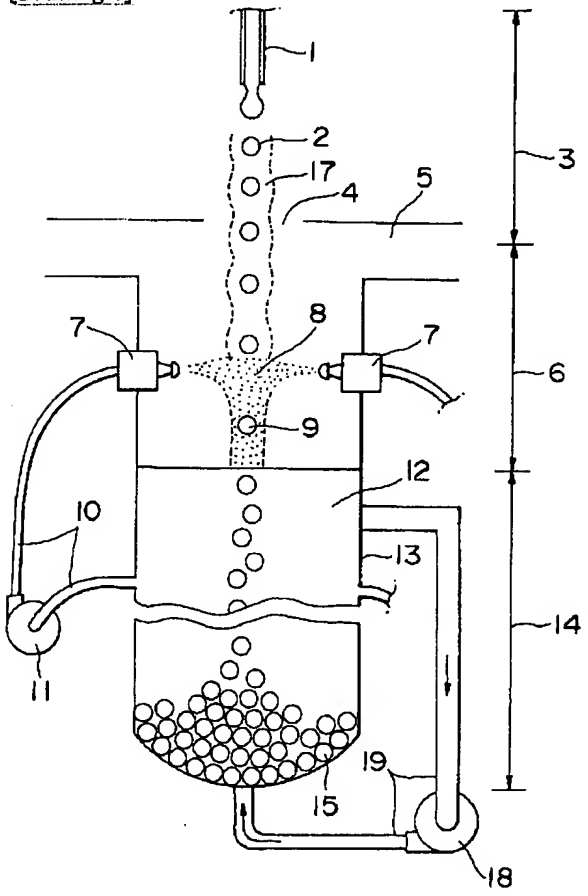
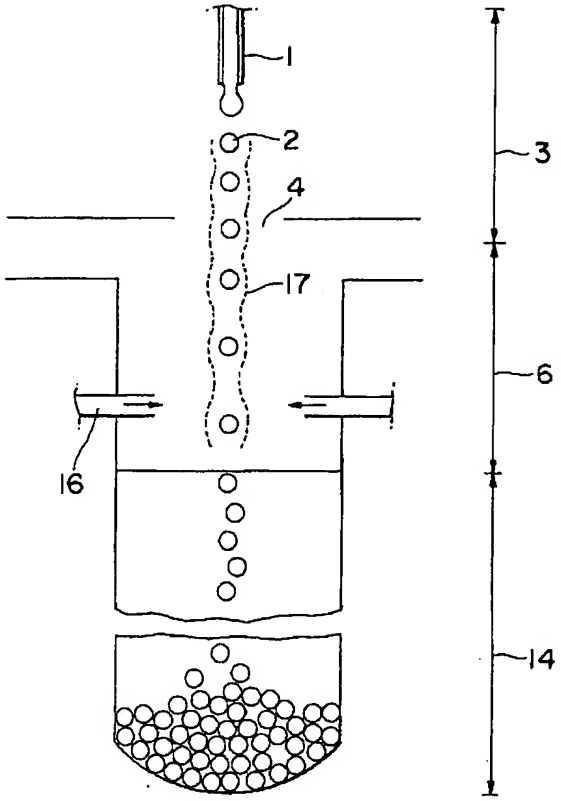
DRAWINGS

[Drawing 1]



[Drawing 2]



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